



Supporting Information

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Uniform and Anisotropic Solid Electrolyte Membrane Enables Superior Solid-State Li Metal Batteries

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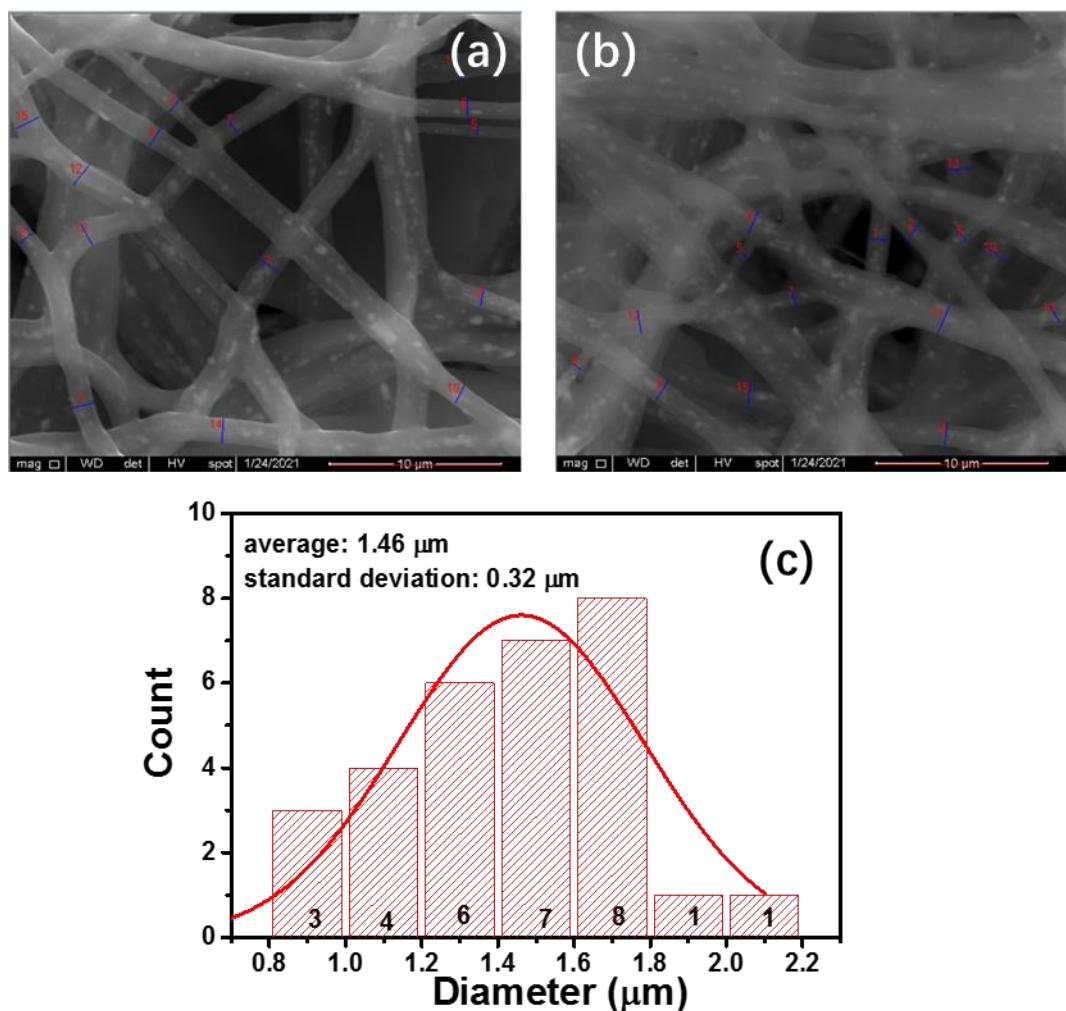


Figure S1. Statistics of the microfiber diameters of ES-CSE.

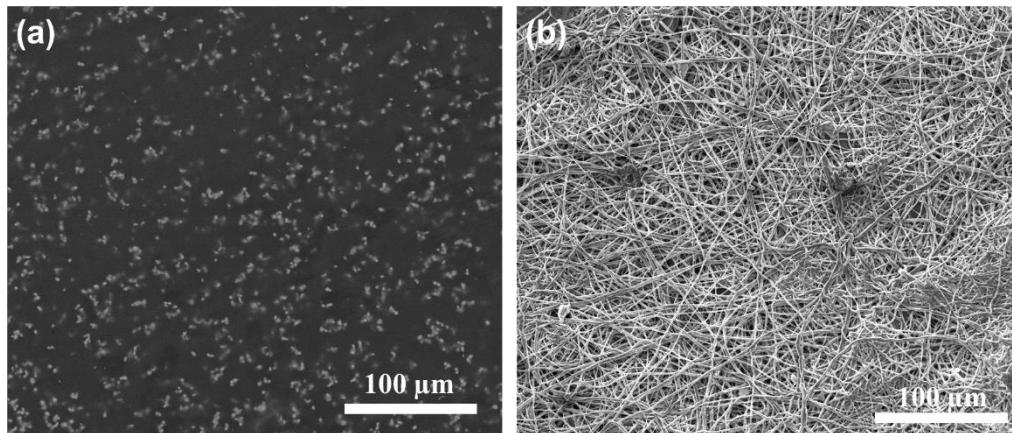


Figure S2. Top-view SEM images of cast-CSE (a) and ES-CSE (b).

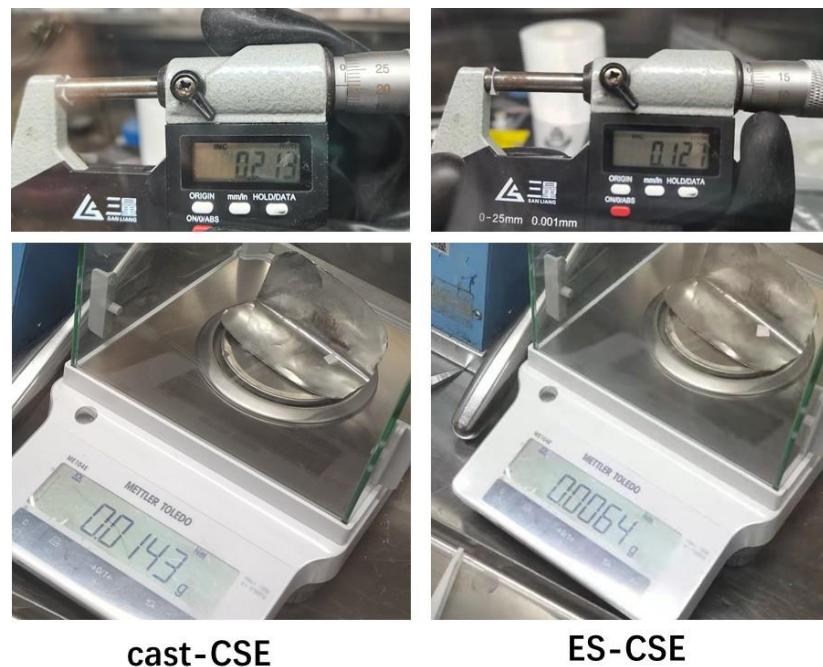


Figure S3. Thicknesses and weights of cast-CSE (a, b) and ES-CSE (c, d) $8 \times 8 \text{ mm}^2$ square membranes.

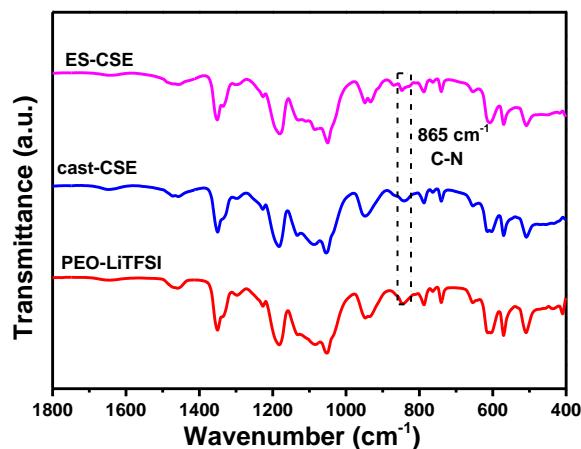


Figure S4. FTIR spectra of PEO-LiTFSI, cast-CSE and ES-CSE.

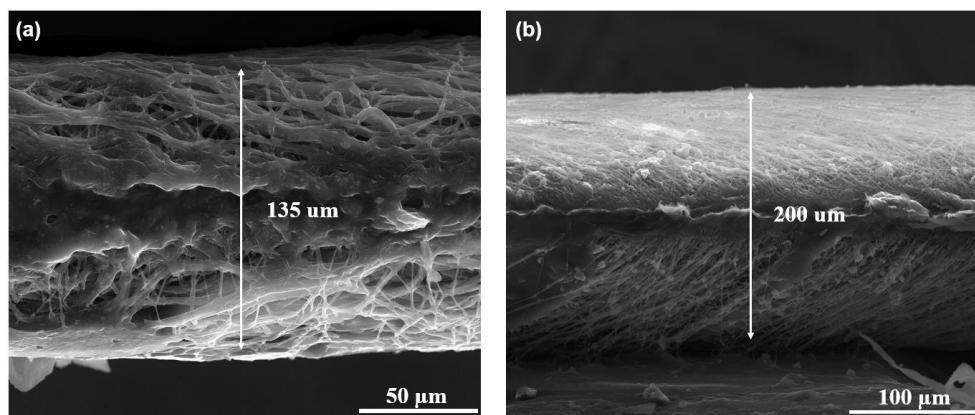


Figure S5. Side-view SEM images of ES-CSE with different electrospinning durations. (a) 5h.

(b) 8h.

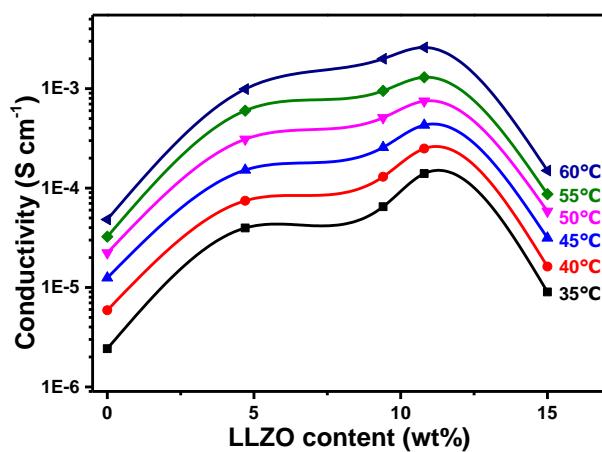


Figure S6. Conductivities of ES-CSE with different LLZO contents at different temperatures.

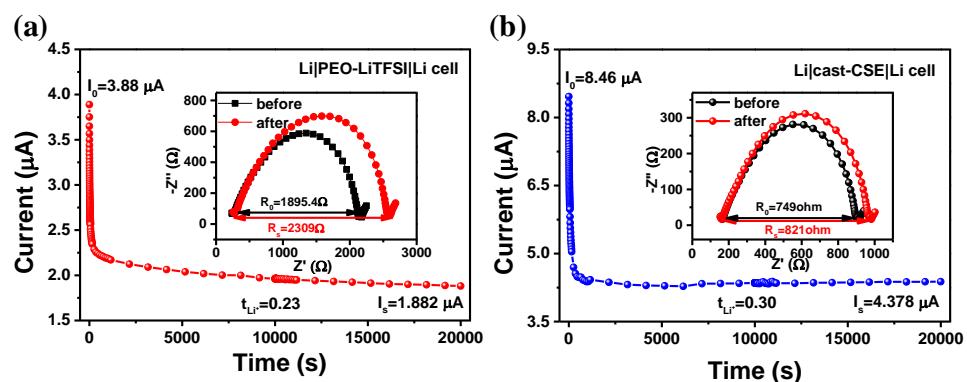


Figure S7. Li-ion transference numbers of PEO-LiTFSI (a) and cast-CSE (b).

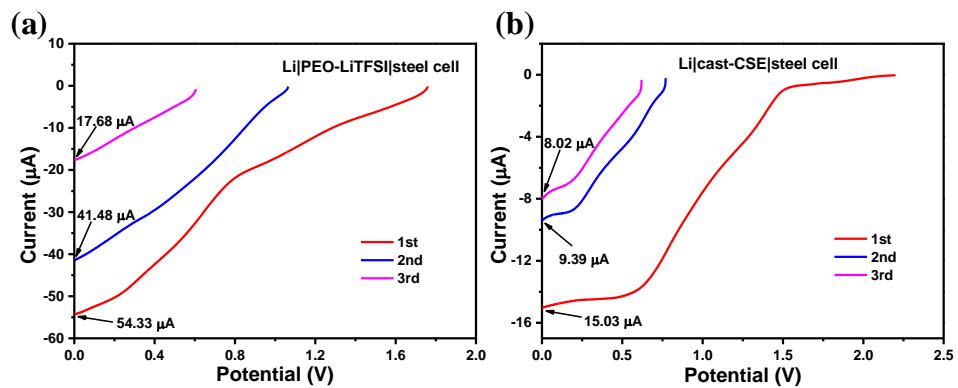


Figure S8. Repeated LSV curves of PEO-LiTFSI (a) and cast-CSE (b) from OCV to 0 V.

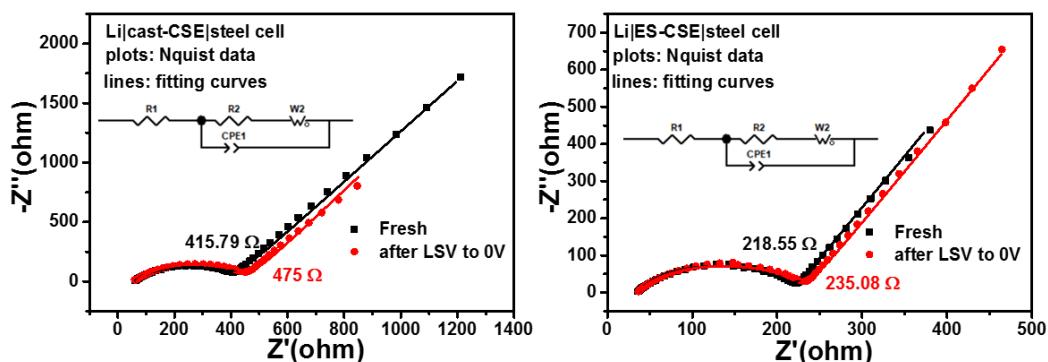


Figure S9. EIS of cast-CSE and ES-CSE based Li||steel cells before and after LSV to 0V.

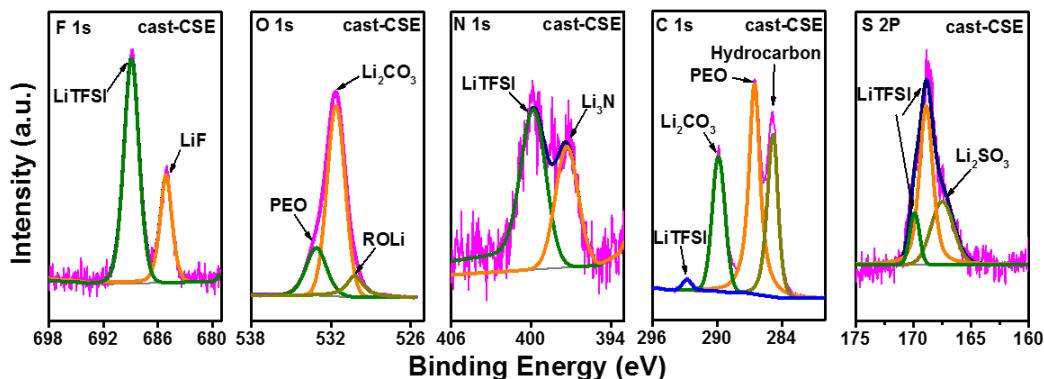


Figure S10. XPS results of the in-situ formed SEI on the steel side for cast-CSE after repeated scanning from OCV to 0 V.

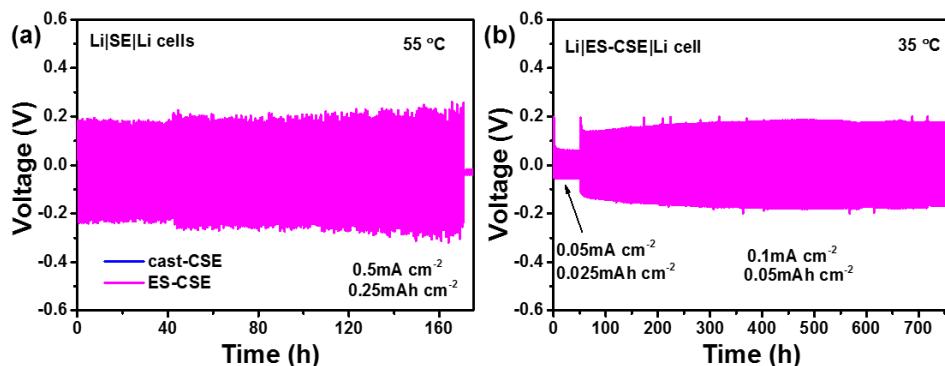


Figure S11. GCD profiles of Li|ES-CSE|Li cell at 0.5 mA cm⁻² and 55 °C (a), 0.1 mA cm⁻² and 35 °C (b).

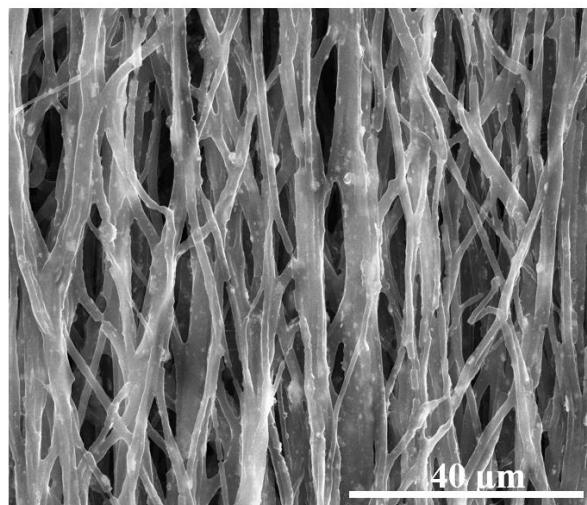


Figure S12. Top-view SEM images of AES-CSE.

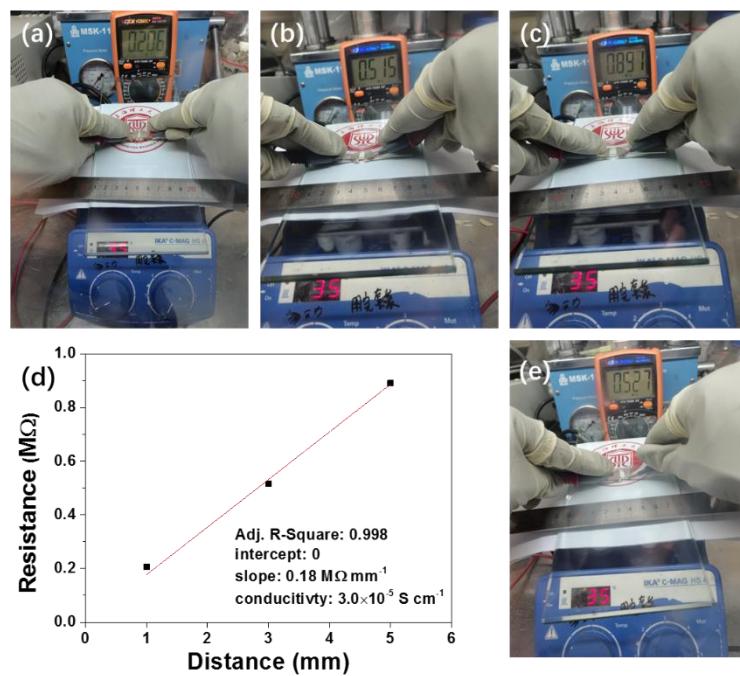


Figure S13. (a, b, c) Optical photographs of the horizontal Li-ion conduction experiments using different distances of 1, 3, and 5 mm. (d) Linear fitting of the resistance versus distance plots. (e) Optical photographs of the horizontal Li-ion conduction experiments using rectangular Li electrodes.

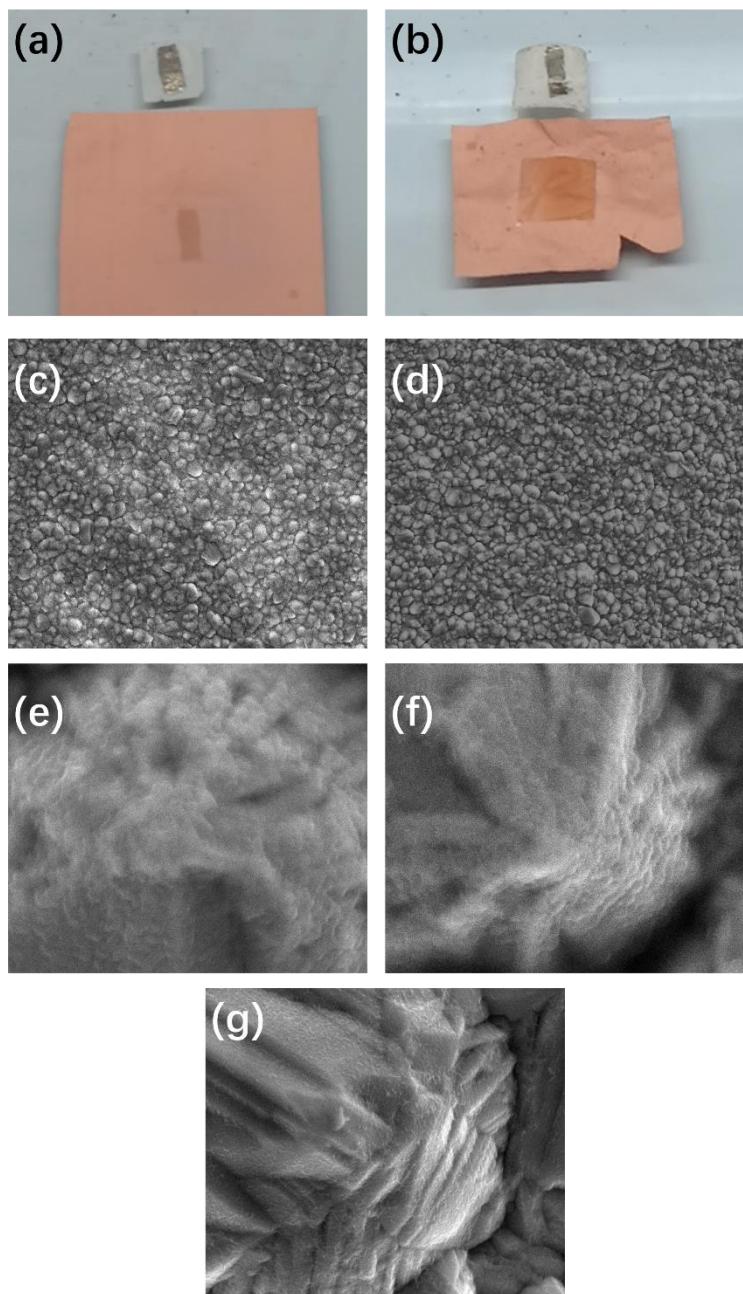


Figure S14. Optical photographs (a, b), SEM images (c, d) and high-resolution SEM images (e, f) of Cu foil after electrochemical Li plating through cast-CSE and ES-CSE. (g) High-resolution SEM image of fresh Cu foil.

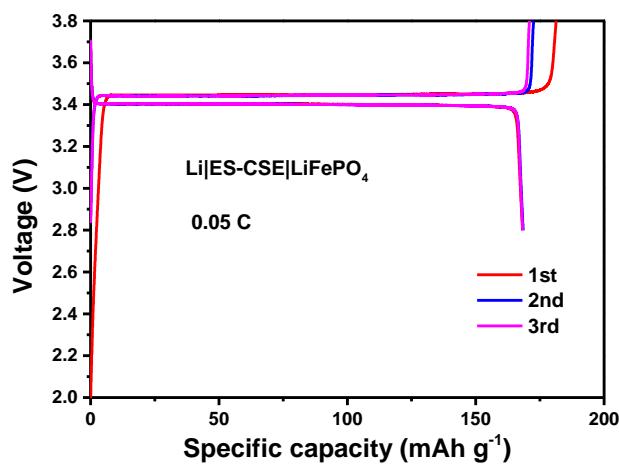


Figure S15. GCD profiles of Li|ES-CSE|LiFePO₄ cell during the activation treatment.

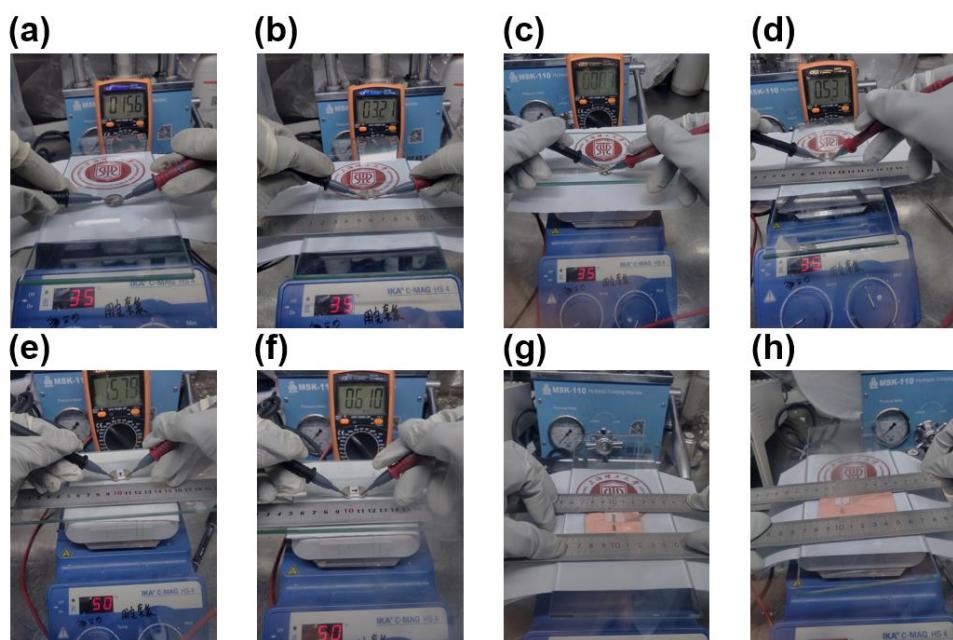


Figure S16. (a-h) Optical photographs of the corresponding vertical/horizontal anisotropic Li-ion conduction experiments.